

Investigations on NO_x reduction by means of laboratory experiments for the release of N-compounds from the fuel bed and simulation of N gas phase reaction kinetics for fiberboard waste

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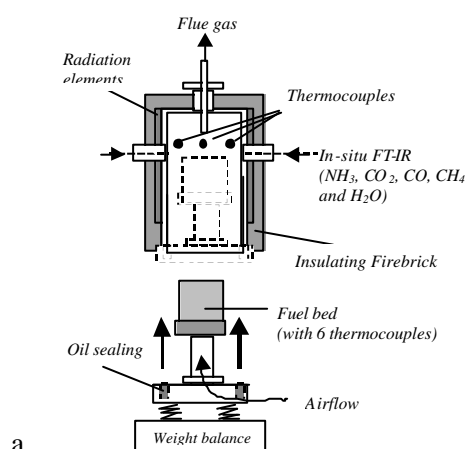
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Introduction

NO_x emissions from biomass combustion are caused by fuel N. The conversion of fuel N to NO_x takes place in two steps: The release of volatile N species (mainly NH₃, HCN, NO) from the fuel bed and their reaction to molecular nitrogen (N₂) or to NO in the gas phase, depending on the reaction conditions. Reducing NO_x emissions by primary measures requires the mechanisms occurring in both steps to be investigated in order to find the global optimum. To do so, experiments to describe the release step and simulations of chemical reaction kinetics have been performed.

Experimental setup and methodology

Laboratory experiments using FT-IR in-situ absorption spectroscopy: For the nitrogen rich fuel fiberboard waste (N-content 3.2 to 4.0 wt%d.b.) test runs have been performed for the investigation of the release of nitrogen compounds from the fuel bed applying a new method of FT-IR in-situ absorption spectroscopy to measure gas concentrations of major components (CO, CO₂, CH₄, NH₃, H₂O) in the hot zone, directly above the fuel bed without extraction of the gas [1].



a.
Figure 1: Laboratory reactor

The laboratory experiments were performed in a cylindrical fixed-bed reactor, with 3500 mm height and 120 mm in diameter. The fuel was put in a cylindrical holder of 100 mm height and 95 mm in inner diameter. Both parts are made of fiberreinforced SiC-ceramics to avoid reactions of CO, NO and ash with the wall and to avoid oxygen entering the reaction zone. The fuel bed is put into the upper part as shown in Figure 1 a. with the dotted lines. Air or a O₂/N₂ mixture is introduced through a porous plate at the bottom of the fuel bed. Test runs have been performed for airflow rates of 20 l/min to 40 l/min and for oxygen concentrations of 8, 13, 16 and 21 Vol% in a O₂/N₂ mixture at a flow rate of 30 l/min. For all experiments the reactor was preheated to 800°C before the sample was entered. Each test was performed in triplicate.

Based on the FT-IR in-situ measurements and the recorded weight loss of the sample, elemental and mass balances have been performed for plausibility tests and the determination of the total release of the elements C, H and N for each experiment in order to prove the applicability of the FT-IR in-situ absorption spectroscopy under hot gas conditions.

Modeling: To investigate the optimum conditions for NO_x reduction, calculations of N-reaction kinetics have been performed for various temperatures of the gas phase in the range of 800°C to 1,200°C and 0 Vol% O₂ to 10 Vol% O₂. Based on the measured gas phase concentrations of the laboratory reactor experiments the reaction kinetics of nitrogen compounds in the gas phase have been investigated with the program codes AURORA and SENKIN from the program package Chemkin 3.6 ® [2] using the GRI 3.0 reaction mechanism.

Results and discussion

Experimental: The experimental results showed a maximum release of TFN (total fixed nitrogen, sum of all N-containing gas species except N_2) at an air flow rate of 25 l/min with a total conversion rate TFN/Fuel-N of 0.58; the combustion time was shortest at 40 l/min and a TFN/Fuel-N rate of 0.28. Reducing the oxygen concentration resulted in longer combustion times, but also decreasing TFN/Fuel-N ratios from 0.48 to 0.33. The major N-species released from the fuel bed during volatilization was NH_3 , which was also expected from literature. Charcoal burnout showed a minor influence on TFN release. The NH_3 release from the fuel bed showed correlation with the CH_4 concentrations in the gas phase and the combustion-specific parameters of the superficial velocity SV (gas Production rate / cross sectional area) and the velocity ratio (ratio of superficial velocity and gas velocity) which forms the base for an empirical modeling. The maximum occurred at a SV of 0.015 m/s.

Modeling: The modeling results showed that in the investigated range the initial concentrations in the gas mixture (CO , CH_4 , CO_2 , H_2O , NH_3) are of minor importance for the NO_x -reduction potential. Temperature and oxygen concentration (stoichiometry) had a major influence at reaction times lower than 0.1 seconds with diverging tendencies for the applied reaction models. For the PFR model higher temperatures and oxygen concentrations showed a faster reaction of NH_3 to NO resulting in higher amounts of NO and radicals within a residence time less than 0.03 sec. In the following NO reduction step increasing temperatures showed a higher NO reduction potential so that for reaction times of 0.1 to 1.0 sec only a small difference of the total TFN reduction potentials can be seen. The optimum stoichiometry turned out to be independent from the applied reactor model (PSR, PFR), but the PSR model showed a faster global reaction and better reduction at high temperatures (1100°C to 1200°C), on the contrary to plug flow conditions (PFR) where the best performance occurred in temperature regions between 900 and 950°C. The highest TFN reduction rates could be achieved with a oxygen concentration of 5 Vol%, representing a oxygen/fuel ratio of approx. 0.64 in the gas phase (TFN/TFN in: 91% at 950°C, PFR; 87% at 1200°C, PSR; both at a reaction time of 0.3 sec).

Conclusions and outlook

The relation of NH_3 release and SV detected opens the base for an empirical modeling of N release from the fuel bed in order to define concentration profiles along a grate. These concentration profiles can be implemented as boundary conditions into CFD-simulations of furnaces as well as in Low NO_x control algorithms of furnaces. For the improvement of this applicability further investigations and experiments have to be performed. To predict NO_x emissions in furnaces for short residence times, flow conditions must be studied in detail, to be able to apply the correct ideal kinetic model. This can be done by comparing residence time distributions calculated by CFD simulations or using the empirical release model as input for CFD NO_x -postprocessors. Special care has to be taken on the reaction model and timescale used in the submodels of the CFD postprocessors. The new application of the FT-IR in-situ absorption spectroscopy proved to be a reliable and valuable tool for hot-gas combustion diagnostics.

References

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